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Serial No.: 09/758,952

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**EXAMINER:** 

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SERIAL NO.:

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FILING DATE:

January 11, 2001

INVENTORS:

Ulrich PEUCHERT and Peter BRIX

TITLE: ALKALI-FREE ALUMINOBOROSILICATE GLASS, AND USES THEREOF

Greensburg, Pennsylvania

Assistant Commissioner for Patents

Washington, D.C. 20231

April 26, 2001

BOX MISSING PARTS

## PRELIMINARY AMENDMENT

Sir:

Please enter the following Preliminary Amendment in order to place the claims of the above-cited application in better condition for examination. Please find attached hereto a markedup version of the changes made to the specification and claims by the current amendment captioned "Version with Markings to Show Changes Made."

# In the Specification:

Please amend the following, without prejudice.

The paragraphs beginning at page 1, line 3, have been amended as follows:

-- This application is related to Application Serial No. 09/758,919, filed concurrently herewith on January 11, 2001, having the title ALKALI-FREE ALUMINOBOROSILICATE GLASS, AND USES THEREOF, naming as inventors Dr. Ulrich PEUCHERT and Dr. Peter NHL:ksm/vwt 1

BRIX, and identified by Attorney Docket No. NHL-SCT-18 US.

This application is also related to Application Serial No. 09/758,946, filed concurrently herewith on January 11, 2001, having the title ALKALI-FREE ALUMINOBOROSILICATE GLASS, AND USES THEREOF, naming as inventors Dr. Ulrich PEUCHERT and Dr. Peter BRIX, and identified by Attorney Docket No. NHL-SCT-20 US.

This application is further related to Application Serial No. 09/758,903, filed concurrently herewith on January 11, 2001, having the title ALKALI-FREE ALUMINOBOROSILICATE GLASS, AND USES THEREOF, naming as inventors Dr. Ulrich PEUCHERT and Dr. Peter BRIX, and identified by Attorney Docket No. NHL-SCT-21 US.--

The paragraph beginning at page 5, line 20, has been amended as follows:

--The glasses described in Japanese Patent Application No. 9-12333 A for hard disks, are comparatively low in Al<sub>2</sub>O<sub>3</sub> or B<sub>2</sub>O<sub>3</sub>, the latter merely being optional. The glasses have high alkaline earth metal oxide contents and have high thermal expansion, which makes them unsuitable for use in LCD or PV technology.--

The paragraphs beginning at page 7, line 4, and ending on page 9, line 7, have been amended as follows:

--The invention teaches that this object can be accomplished by aluminoborosilicate glasses having a coefficient of thermal expansion  $\alpha_{20/300}$  of between 2.8 x  $10^{-6}/K$  and 3.8 x  $10^{-6}/K$ , which has the following composition (in % by weight, based on oxide):

silicon dioxide (SiO<sub>2</sub>) - from somewhat more than 58% to 65% (> 58% - 65%); boric oxide (B<sub>2</sub>O<sub>3</sub>) - from somewhat more than 6% to 10.5% (> 6% - 10.5%); aluminum oxide (Al<sub>2</sub>O<sub>3</sub> - from somewhat more than 14% to 25% (> 14% - 25%); magnesium oxide (MgO) from 0% to somewhat less than 3% (0 - < 3%); calcium oxide (CaO) - from 0% to 9% (0% - 9%); strontium oxide (SrO) - from 0.1% to 1.5% (0.1% - 1.5%); barium oxide (BaO) - from somewhat more than 5% to 8.5% (> 5% - 8.5%); with strontium oxide (SrO) + barium oxide (BaO) - equal to or somewhat less than 8.6% ( $\leq$  8.6%); and with magnesium oxide (MgO) + calcium oxide (CaO) + strontium oxide (SrO) + barium oxide (BaO) - from 8% to 18% (8% - 18%); and zinc oxide

(ZnO) - from 0% to somewhat less than 2% (0% - < 2%).

The invention also teaches an alkali-free aluminoborosilicate glass having a coefficient of thermal expansion  $\alpha_{20/300}$  of between 2.8 x  $10^{-6}$ /K and 3.6 x  $10^{-6}$ /K, which has the following composition (in % by weight, based on oxide): silicon dioxide (SiO<sub>2</sub>) - from somewhat more than 58% to 64.5% (> 58% - 64.5%); boric oxide (B<sub>2</sub>O<sub>3</sub>) - from somewhat more than 6% to 10.5% (> 6% - 10.5%); aluminum oxide (Al<sub>2</sub>O<sub>3</sub>) - from 20.5% to 24% (20.5% - 24%); magnesium oxide (MgO) - from 0% to somewhat less than 3% (0% - < 3%); calcium oxide (CaO) - from 2.5% to somewhat less than 8% (2.5% - < 8%); strontium oxide (SrO) - from 0.1% to 3.5% (0.1% - 3.5%); barium oxide (BaO) - from somewhat more than 5% to 7.5% (> 5% - 7.5%); with strontium oxide (SrO) +

barium oxide (BaO) being equal to or less than 8.6% ( $\leq 8.6\%$ ); and with magnesium oxide (MgO) + calcium oxide (CaO) + strontium oxide (SrO) + barium oxide (BaO) in the range of from 8% to 18% (8% - 18%); and zinc oxide (ZnO) - from 0% to somewhat less than 2% (0% - < 2%).

The glass contains between > 58 and 65% by weight of SiO<sub>2</sub>. At lower contents, the chemical resistance is impaired, while at higher levels, the thermal expansion is too low and the crystallization tendency of the glass increases. Preference is given to a maximum content of 64.5% by weight.

The glass contains relatively high levels of  $(Al_2O_3)$ , i.e. > 14 - 25% by weight of  $(Al_2O_3)$ , preferably at least 18% by weight, particularly preferably > 18% by weight. These relatively high  $Al_2O_3$  levels are favourable to the crystallization stability of the glass and have a positive effect on its heat resistance without excessively increasing the processing temperature. Particular preference is given to a content of at least 20.5% by weight, most preferably of at least 21.5% by weight, of  $(Al_2O_3)$ . Preference is given to a maximum  $Al_2O_3$  content of 24% by weight.

The  $B_2O_3$  content is > 6 - 10.5% by weight. The  $B_2O_3$  content is restricted to the maximum content specified in order to achieve a high glass transition temperature  $T_g$ . Higher contents would also impair the chemical resistance to hydrochloric acid solutions. (Preference is given to a maximum  $B_2O_3$  content of 11%

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by weight). The minimum  $B_2O_3$  content specified serves to ensure that the glass has good meltability and good crystallization stability. Preference is given to a minimum content of > 8% by weight of B,O,.

The network-forming components (Al<sub>2</sub>O<sub>3</sub>) and B<sub>2</sub>O<sub>3</sub> are preferably present at mutually dependent minimum levels, ensuring a sufficient content of the network formers SiO2, (Al2O3) and B2O3. For example, in the case of a  $B_2O_3$  content of > 6 - 10.5% by weight, the minimum Al<sub>2</sub>O<sub>3</sub> content is preferably > 18% by weight, and in the case of an (Al<sub>2</sub>O<sub>3</sub>) content of > 14 - 25% by weight, the minimum  $B_2O_3$  content is preferably > 8% by weight. Preferably, in particular in order to achieve low thermal expansion coefficients of up to 3.6 x  $10^{-6}/K$ , the sum of  $SiO_2$ ,  $B_2O_3$ and Al<sub>2</sub>O<sub>3</sub> is at least 85% by weight.--

The paragraph beginning at page 10, line 6, has been amended as follows:

--On the other hand, the maximum SrO content can be up to 3.5% by weight in the case of high-Al2O3 (in particular > 20.5% by weight) and relatively CaO-rich (in particular > 2.5% by weight) glasses. The higher SrO content has the positive effect of counteracting the slight increase in crystallization tendency found in relatively CaO-rich glasses having relatively high Al2O3 contents. --

The paragraph beginning at page 11, line 10, has been

amended as follows:

--In addition to the low-SrO glass of the main claim, a glass having the desired requirement profile and a coefficient of thermal expansion  $\alpha_{20/300}$  of between 2.8 x  $10^{-6}/K$  and 3.6 x  $10^{-6}/K$  is also described by the following composition (in % by weight, based on oxide):  $SiO_2 > 58 - 64.5$ ,  $B_2O_3 > 6 - 10.5$ ,  $Al_2O_3 > 20.5 - 24$ , MgO 0 - < 3, CaO 2.5 - < 8, SrO 0.1 - 3.5 and BaO > 5 - 7.5, with SrO + BaO  $\leq$  8.6 and with MgO + CaO + SrO + BaO 8 - 18; ZnO 0 - < 2.--

The paragraph beginning at page 12, line 5, has been amended as follows:

--The glass may contain conventional refining agents in the usual amounts: it may thus contain up to 1.5% by weight of As<sub>2</sub>O<sub>3</sub>, Sb<sub>2</sub>O<sub>3</sub>, SnO<sub>2</sub>, CeO<sub>2</sub>, Cl<sup>-</sup> (for example in the form of BaCl<sub>2</sub>), F<sup>-</sup> (for example in the form of CaF<sub>2</sub>) and/or SO<sub>4</sub><sup>2-</sup> (for example in the form of BaSO<sub>4</sub>). The sum of the refining agents should, however, not exceed 1.5% by weight. If the refining agents As<sub>2</sub>O<sub>3</sub> and Sb<sub>2</sub>O<sub>3</sub> are omitted, the glass can be processed not only using a variety of drawing methods, but also by the float method.--

The portion of the table located on page 16 has been amended as follows:

	7	8	9	10	11	12
SiO <sub>2</sub>	58.2	58.1	60.5	61.5	62.0	61.0
B <sub>2</sub> O <sub>3</sub>	7.6	7.6	9.5	9.6	9.5	6.2
Al <sub>2</sub> O <sub>3</sub>	21.4	21.5	18.2	17.1	16.5	18.5
MgO	2.8	2.8	1.9	1.9	2.7	1.0
Ca0	2.5	2.5	2.6	2.6	1.3	6.0
Sr0	2.0	1.0	1.0	0.5	0.7	1.0
BaO	5.2	5.2	6.0	6.5	7.0	5.5
ZnO		1.0	_	_	_	0.5
$\alpha_{20/300}$ [10 <sup>-6</sup> /K]	3.18	3.09	3.04	3.04	3.02	3.46
ρ [g/cm³]	2.51	5.52	2.46	2.44	2.48	2.53
T <sub>g</sub> ['C]	747	742	727	723	715	740
T4 [°C]	1303	1305	1320	1325	1309	1315
T2 [°C]	1655	1660	1671	1678	1681	n.m.
$n_d$	1.522	1.522	1.514	1.512	1.510	n.m.
HC1 [mg/cm²]	n.m.	n.m.	n.m.	n.m.	n.m.	n.m.
BHF [mg/cm²]	0.65	0.64	0.53	0.50	0.52	n.m.

The portion of the table located on page 17 has been amended as follows:

	13	14	15	16
SiO <sub>2</sub>	59.9	58.9	59.9	59.7
$B_2O_3$	8.5	8.5	6.5	8.0
Al <sub>2</sub> O <sub>3</sub>	15.5	16.5	18.5	20.5
MgO	2.0	0.6	2.8	1.6
CaO	7.2	8.2	6.0	2.5
SrO	1.0	0.5	0.5	2.0
BaO	5.1	5.5	5.5	5.1
ZnO	0.5	1.0	_	_
$\alpha_{20/300}$ [10 <sup>-6</sup> /K]	3.74	3.75	3.57	3.03
ρ [g/cm³]	2.52	2.53	2.53	2.495
T <sub>g</sub> [°C]	706	708	737	740
T4 ['C]	1264	1266	1291	1324
T2 ['C]	1623	1624	1646	1708
$n_d$	1.524	1.526	1.526	1.517
HCl [mg/cm <sup>2</sup> ]	0.38	0.37	0.27	0.99
BHF [mg/cm <sup>2</sup> ]	0.53	0.51	0.58	0.59

The paragraph beginning at page 18, line 26, has been amended as follows:

--The glasses have high thermal shock resistance and good devitrification stability. The glasses can be produced as flat glasses by the various drawing methods, for example microsheet

down-draw, up-draw or overflow fusion methods, and, in a preferred embodiment, if they are free from  $As_2O_3$  and  $Sb_2O_3$ , also by the float process.--

The paragraph beginning at page 21, line 18, has been amended as follows:

--Another feature of the invention resides broadly in an aluminoborosilicate glass, characterized in that it comprises at least 18% by weight, preferably more than 18% by weight, of  $Al_2O_3$ .--

The paragraph beginning at page 21, line 28, and ending on page 22, line 3, has been amended as follows:

--A further feature of the invention resides broadly in an alkali-free aluminoborosilicate glass having a coefficient of thermal expansion  $\alpha_{20/300}$  of between 2.8 x  $10^{-6}/K$  and 3.6 x  $10^{-6}/K$ , which has the following composition (in % by weight, based on oxide):  $SiO_2 > 58 - 64.5$ ;  $B_2O_3 > 6 - 10.5$ ;  $Al_2O_3 = 20.5 - 24$ ; MgO 0 - < 3; CaO 2.5 - < 8; SrO 0.1 - 3.5; BaO > 5 - 7.5; with SrO + BaO  $\leq 8.6$ ; with MgO + CaO + SrO + BaO  $\leq 8.6$ ; with MgO + CaO + SrO + BaO  $\leq 8.6$ ; ZnO 0 - < 2.--

The paragraph beginning at page 22, line 13, has been amended as follows:

--A further feature of the invention resides broadly in an aluminoborosilicate glass, characterized in that it additionally comprises:  $ZrO_2$  0 - 2;  $TiO_2$  0 - 2; with  $ZrO_2$  +  $TiO_2$  0 - 2;  $As_2O_3$  0 - 1.5;  $Sb_2O_3$  0 - 1.5;  $SnO_2$  0 - 1.5;  $CeO_2$  0 - 1.5;  $C1^-$  0 - 1.5;  $F^-$  0

- 1.5;  $SO_4^{2-}$  0 - 1.5; with  $As_2O_3 + Sb_2O_3 + SnO_2 + CeO_2 + Cl^- + F^- + <math>SO_4^{2-} \le 1.5.$ 

The paragraphs beginning at page 23, line 25, and ending on page 24, line 26, have been amended as follows:

--The corresponding foreign and international patent publication applications, namely, Federal Republic of Germany Patent Application No. 100 00 836.4-45, filed on January 12, 2000, having inventors Dr. Ulrich PEUCHERT and Dr. Peter BRIX, as well as their published equivalents, and other equivalents or corresponding applications, if any, in corresponding cases in the Federal Republic of Germany and elsewhere, and the references cited in any of the documents cited herein, are hereby incorporated by reference as if set forth in their entirety herein, are hereby incorporated by reference as if set forth in their entirety herein, are hereby herein.

The corresponding foreign and international patent publication applications, namely, Federal Republic of Germany Patent Application No. 100 00 839.9-45, filed on January 12, 2000, having inventors Dr. Ulrich PEUCHERT and Dr. Peter BRIX, as well as their published equivalents, and other equivalents or corresponding applications, if any, in corresponding cases in the Federal Republic of Germany and elsewhere, and the references cited in any of the documents cited herein, are hereby incorporated by reference as if set forth in their entirety

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herein, are hereby incorporated by reference as if set forth in their entirety herein.

The corresponding foreign and international patent publication applications, namely, Federal Republic of Germany Patent Application No. 100 00 837.2-45, filed on January 12, 2000, having inventors Dr. Ulrich PEUCHERT and Dr. Peter BRIX, as well as their published equivalents, and other equivalents or corresponding applications, if any, in corresponding cases in the Federal Republic of Germany and elsewhere, and the references cited in any of the documents cited herein, are hereby incorporated by reference as if set forth in their entirety herein, are hereby incorporated by reference as if set forth in their entirety herein .--

# In the Claims:

Please rewrite the following claims, without prejudice. --1. (Amended) Alkali-free aluminoborosilicate glass having a coefficient of thermal expansion  $\alpha_{\rm 20/300}$  of between 2.8 x  $10^{-5}/K$  and 3.8 x  $10^{-6}/\text{K}$ , which has the following composition (in % by weight, based on oxide):

SiO <sub>2</sub>	> 58 - 65
$B_2O_3$	> 6 - 10.5
$Al_2O_3$	> 14 - 25
MgO	0 - < 3
CaO	0 0

SrO	0.1 - 1.5
BaO	> 5 - 8.5
with SrO + BaO	≤ 8.6
with MgO + CaO + SrO + BaO	8 - 18
ZnO	0 - < 2.

- 2. (Amended) Aluminoborosilicate glass according to Claim 1, characterized in that it comprises at least 18% by weight, preferably more than 18% by weight, of  $Al_2O_3$ .
- 3. (Amended) Aluminoborosilicate glass according to Claim 2, characterized by the following composition (in % by weight, based on oxide):

SiO <sub>2</sub>	> 58 - 64.5
$B_2O_3$	> 6 - 10.5
$Al_2O_3$	> 18 - 24
MgO	0 - < 3
Ca0	1 - < 8
SrO	0.1 - 1.5
BaO	> 5 - 8
with SrO + BaO	< 8.5
with MgO + CaO + SrO + BaO	8 - 18
ZnO	0 - < 2.

4. (Amended) Aluminoborosilicate glass according to Claim 3, characterized in that it comprises at least 20.5% by weight of  $Al_2O_3$ .

5. (Amended) Alkali-free aluminoborosilicate glass having a coefficient of thermal expansion  $\alpha_{20/300}$  of between 2.8 x  $10^{-6}/K$  and 3.6 x  $10^{-6}/K$ , which has the following composition (in % by weight, based on oxide):

SiO <sub>2</sub>	> 58 - 64.5
$B_2O_3$	> 6 - 10.5
Al <sub>2</sub> O <sub>3</sub>	20.5 - 24
MgO	0 - < 3
CaO	2.5 - < 8
SrO	0.1 - 3.5
BaO	> 5 - 7 5
with SrO + BaO	≤ 8.6
with MgO + CaO + SrO + BaO	8 - 18
ZnO	0 - < 2.

- 6. (Amended) Aluminoborosilicate glass according to Claim 5, characterized in that it comprises at least 21.5% by weight of  ${\rm Al_2O_3}$ .
- 7. (Amended) Aluminoborosilicate glass according to Claim 6, characterized in that it comprises more than 8% by weight of  $B_2O_3$ .
- 8. (Amended) Aluminoborosilicate glass according to Claim 7, characterized in that it comprises at least 0.1% by weight of ZnO.
- 9. (Amended) Aluminoborosilicate glass according to Claim 8,

characterized in that it additionally comprises:

ZrO <sub>2</sub>	0 - 2
$\mathtt{TiO}_2$	0 - 2
with ZrO <sub>2</sub> + TiO <sub>2</sub>	0 - 2
$As_20_3$	0 - 1.5
$Sb_2O_3$	0 - 1.5
SnO <sub>2</sub>	0 - 1.5
CeO <sub>2</sub>	0 - 1.5
C1 <sup>-</sup>	0 - 1.5
F <sup>-</sup>	0 - 1.5
SO <sub>4</sub> <sup>2-</sup>	0 - 1.5
with $As_2O_3 + Sb_2O_3 + SnO_2 + CeO_2$	≤ 1.5
+ Cl <sup>-</sup> + F <sup>-</sup> + SO <sub>4</sub> <sup>2-</sup> .	

- 10. (Amended) Aluminoborosilicate glass according to Claim 9, characterized in that it is free of arsenic oxide and antimony oxide, apart from unavoidable impurities, and that it can be produced in a float plant.
- 11. (Amended) Aluminoborosilicate glass according to Claim 10, which has a coefficient of thermal expansion  $\alpha_{20/300}$  of 2.8 x  $10^{-6}/K$  3.6 x  $10^{-6}/K$ , a glass transition temperature Tg of > 700°C and a density  $\rho$  of < 2.600 g/cm<sup>3</sup>.
- 12. (Amended) Use of the aluminoborosilicate glass according to Claim 1 as substrate glass in display technology.
- 13. (Amended) Use of the aluminoborosilicate glass according to

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Claim 1 as substrate glass in thin-film photovoltaics.--

### REMARKS

This Preliminary Amendment is being submitted in order to place the present application in a better condition for examination. Specifically, numerous typographical errors in the specification and claims have been corrected herein. Further, the claims have been amended to remove the multiple dependencies. Care has been taken to avoid the introduction of new matter.

None of the changes submitted in this Preliminary Amendment are to be construed as having any effect on the scope of the subject matter being claimed, as they are merely being presented for clarification purposes. All of the changes made in this Preliminary Amendment are made without prejudice, so that the matter deleted may be reintroduced as necessary for prosecution of the application.

## Summary and Conclusion:

It is submitted that Applicants have provided a new and unique ALKALI-FREE ALUMINOBOROSILICATE GLASS, AND USES THEREOF. It is submitted that the claims, as amended, are fully distinguishable over the prior art. Therefore, it is requested that a Notice of Allowance be issued at an early date.

If mailed, I, the person signing this certification below, hereby certify that this correspondence is being deposited with the United States Postal Service with sufficient postage as first

class mail in an envelope addressed to: Assistant Commissioner for Patents, Washington, D.C. 20231, on the date indicated in the certification of mailing on the transmittal letter sent herewith, or if facsimile transmitted, I, the person signing this certification below, hereby certify that this paper is being facsimile transmitted to the United States Patent and Trademark Office on the date indicated in the certification of facsimile transmission on the transmittal letter which is being facsimile transmitted herewith.

Respectfully submitted,

Nils H. Ljungman, Esq.

Attorney for Applicant[s] Reg. No. 25,997

Name of person signing certification

Nils H. Ljungman & Associates

P.O. Box 130

Greensburg, PA 15601-0130 Telephone: (724) 836-2305

Facsimile: (724) 836-2313